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14. ABSTRACT
The most significant accomplishments in this project are: (a) the demonstration of optically pumped silicon based light emitters utilizing colloidal PbSe QD which are inserted in PC microcavities for efficient coupling. Enhancements of spontaneous emission with a linewidth of ~2.0 nm, corresponding to a cavity Q factor of 775, is observed at 1550 nm at room temperature; (b) an electrically injected silicon based light source using PbSe QDs, which are more compact and versatile. With a current density of 113 mA/cm2, a resonance at λ=1669 nm having a linewidth of 4nm is observed, which corresponds to a cavity Q factor of ~420. This nanoscale light source based on silicon, which is capable of being fabricated on CMOS chips, is of interest as a practical technology for optical interconnects in silicon photonics; (c) the demonstration of possibility of surface Plasmon enabled nanolaser with round-trip gain in the sense that the transmission in the waveguide increases as the pumping power increases.

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Significant Accomplishments:

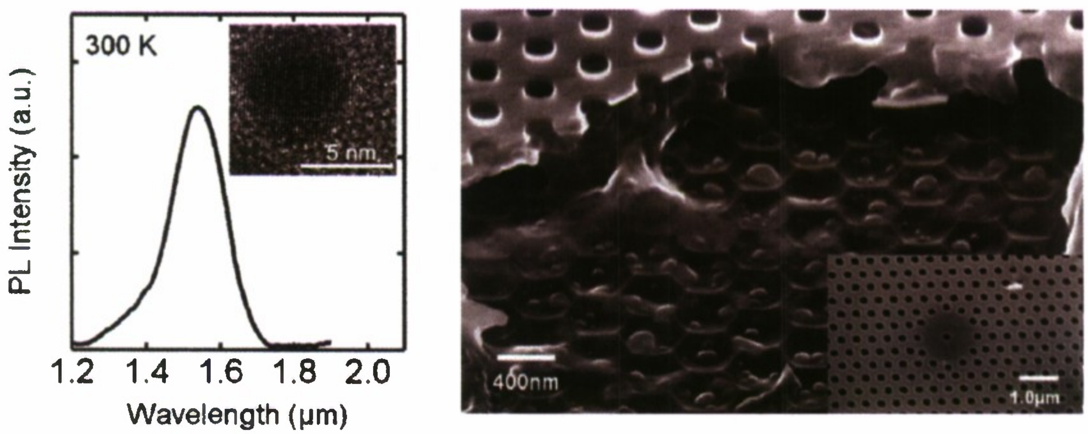
The most significant accomplishments in this project are : (a) the demonstration of optically pumped silicon based light emitters utilizing colloidal PbSe QDs which are inserted in PC microcavities for efficient coupling. Enhancement of spontaneous emission with a linewidth of ~2.0 nm, corresponding to a cavity Q factor of 775, is observed at 1550 nm at room temperature; (b) an electrically injected silicon based light source using PbSe QDs, which are more compact and versatile. With a current density of 113 mA/cm^2, a resonance at λ=1669 nm having a linewidth of 4 nm is observed, which corresponds to a cavity Q factor of ~420. This nanoscale light source based on silicon, which is capable of being fabricated on CMOS chips, is of interest as a practical technology for optical interconnects in silicon photonics; (c) the demonstration of possibility of surface plasmon enabled nanolaser with round-trip gain in the sense that the transmission in the waveguide increases as the pumping power increases. A more detailed description of these accomplishments follows.

1. Enhanced spontaneous emission from colloidal PbSe quantum dots in silicon-based photonic crystal microcavities

A silicon-based nanoscale light source which operates at the wavelength of ~1.55 μm is highly desirable for future high-speed circuits and systems. The most promising approach to realize it is to use chemically synthesized nanocrystal, such as PbSe colloidal quantum dots (QDs), as a gain medium. By placing the emissive QDs in a high-Q photonic crystal (PC) microcavity, wherein light strongly confined in a modal volume of the order of a cubic optical wavelength, the dynamics of spontaneous emission from the dots can be modified due to the Purcell effect.

PbSe QDs were synthesized using a noncoordinating solvent technique. The synthesis procedure starts with the preparation of lead oleate solution from PbO and oleic acid. The injection of selenium-trioctylphosphine reagents into the reaction solution at an elevated temperature (~160 °C) induces the nucleation of PbSe and the subsequent cooling down to 135 °C allows the nuclei to grow into highly crystalline nanoparticles. By carefully controlling the growth conditions such as the growth temperature, injection, and nanocrystal growth time, hghly monidisperse PbSe QDs can be produced without any further size-selective precipitation process. The resulting PbSe QDs were stabilized with a capping layer of oleate molecules coordinated to the Pb atoms. Shown in the inset of Fig. 1(a) is the high resolution cross-sectional transmission electron microscopy image of a single PbSe QD with a diameter of ~6 nm. The PbSe nanocrystals are dispersed in tetrachloroethylene for photoluminescence (PL) measurement. The PbSe QDs exhibit strong emission peak at ~1.55 μm at room temperature, as shown in Fig. 1(a).

Silicon two-dimensional PC microcavities were fabricated on silicon-on-insulator (SOI) wafers, which consist of 0.23 μm Si atop 0.4 μm SiO_2. The PC, consisting of a triangular lattice of air holes with period a=425 nm and hole radius r=125 nm, was fabricated by electron beam lithography and electron cyclotron resonance reactive ion etching with silicon oxide used as an etch mask. The PC air holes were etched completely through the top silicon layer. A freestanding Si slab was obtained by selectively etching away the underlying SiO_2 layer with buffered hydrofluoric acid. The scanning electron microcope (SEM) image of a H2 photonic crystal microcavity is shown in Fig. 1(b). The air holes surrounding the cavity were shifted outward and reduced in size to improve the Q factor. A smaller air hole (~250 nm in diameter) was incorporated at the center of the H2 microcavity. To insert PbSe QDs into the air holes, the Si PC was soaked in the PbSe QD solution for 10 h, followed by a quick rinse in de-ionized water. The QDs that attach to the nanohole at the center of the PC microcavity serve as the gain medium.



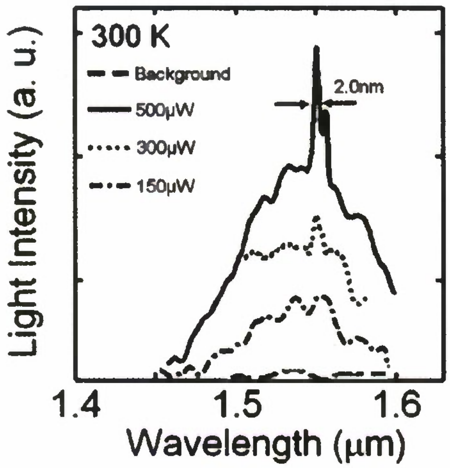
(a) (b)

Figure 1 (a) Room-temperature photoluminescence spectrum of colloidal PbSe quantum dots with inset showing high resolution cross-sectional transmission electron microscope image of a single quantum dot; (b) scanning electron microscope image of the fabricated Si two-dimensional photonic crystal microcavity, and of a cleaved section of the photonic crystal showing PbSe quantum dots attached inside the air holes and on the silicon substrate below the air gap.

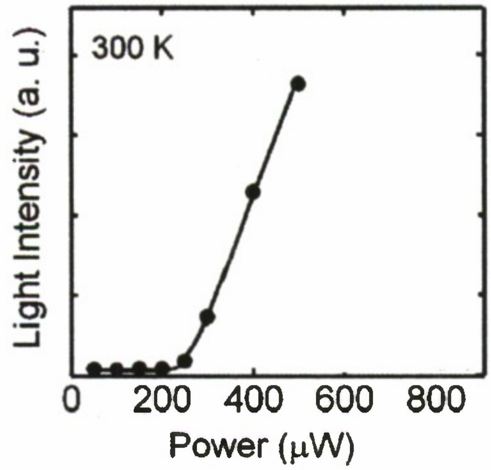
The presence of an air hole at the cavity center modifies the cavity modes considerably. Using three-dimensional finite difference time domain (FDTD) techniques, the modified mode profile, which is shifted outward due to the presence of the air hole, is observed. The presence of PbSe QDs in the air holes of the PC during the soaking process, as seen in the SEM image of cleaved sections of the PC of Fig. 1(b), reduces the quality factor. With an estimated fill factor of the dots in the holes, resulting in a modified index of 1.6 therein, we calculate a microcavity Q of ~900–1100.

The PC microcavities with embedded PbSe QDs were optically excited at room temperature with a Ti:saphire laser operating at 830 nm. Emission from the microcavity was focused with an infinity-corrected 100 × objective lens (numerical aperture=0.7) and analyzed by a 0.75 m high resolution spectrometer. The output of the spectrometer was detected with an InGaAs photomultiplier tube using lock-in amplification. The output spectral characteristics of a H2 microcavity under continuous wave (cw) excitation at different pump powers are shown in Fig. 2(a). Two nearly degenerate dipole modes at 1550.6 and 1556.3 nm, respectively are clearly observed at elevated pump powers. The slight splitting in the mode energies is likely due to imperfections in the fabricated photonic crystal structures. The emission with peak at ~1550.6 nm exhibits a linewidth of ~2.0 nm, corresponding to a cavity Q factor of ~775. The slightly lower value of Q, compared to the estimated one, is attributed to imperfections in the fabrication process and heating effect during measurement.

It may also be noted that the output spectrum consists of a relatively large background, which is believed to be due to contributions from PbSe QDs residing in the PC air holes as well as those beneath the photonic crystal membrane. The light-light characteristics of the H2 microcavity, shown in Fig. 2(b), are derived from the integrated cavity resonance intensity by subtracting the background emission for varying excitation power levels. The turn on at ~230 μW does not indicate onset of laser action, but simply represents the power at which the cavity resonance peak intensity becomes larger than that of the background luminescence. From three-dimensional FDTD simulation, the mode volume is calculated to be $\sim 1.8(\lambda_c/n)^3$, and using Q=775 we estimate a Purcell factor of ~35.



(a)

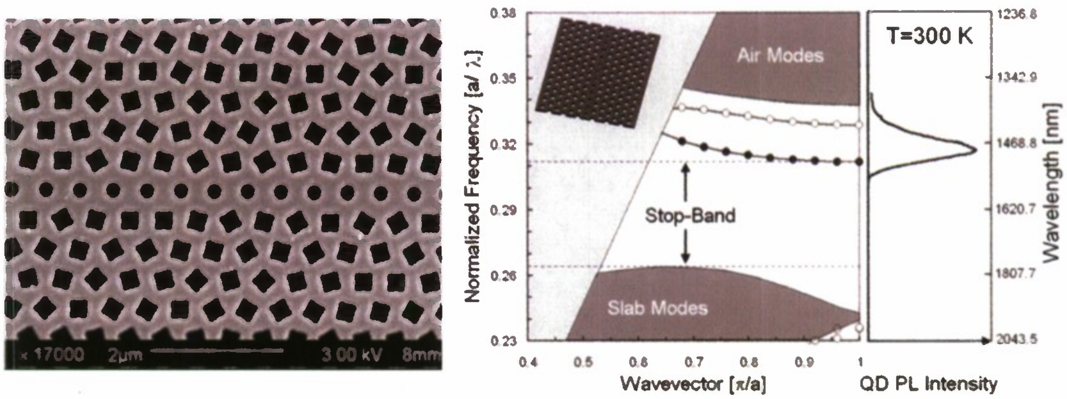


(b)

Figure 2 (a) Output spectra of the H2 microcavity with PbSe quantum dots measured at 300 K for different pump powers. The background spectrum is recorded from a region of the wafer where there is no photonic crystal with air holes; (b) measured light-light characteristics of the dipole mode at 1550.6 nm.

2. Enhanced photoluminescence from embedded colloidal PbSe quantum dots in silicon-based random photonic crystal microcavities

PCs are periodic dielectric structures, usually two-dimensional (2D) arrays of air holes in high-refractive-index membranes, which selectively inhibit light propagation in certain bands of frequencies. Destroying the periodicity of the lattice introduces small defects which act as optical cavities with high Qs wherein light can be localized by total internal and Bragg reflections. Q factors of the order of 10^6 have been measured in engineered microcavities in 2D PCs. On the other hand, a different approach to photon localization in PCs have recently investigated and reported, which relies on random structural perturbations introduced uniformly throughout the crystal by deliberately changing the shapes and orientations of the lattice elements (air holes). Such random disorder superimposed onto the crystal causes backscattering which impedes propagation of Bloch waves along line defects defined in the 2D lattice. Extended modes that propagate with a low group velocity at frequencies approaching the mode edge become spatially confined in sections of the disordered waveguide. This subtle interplay of order and disorder was predicted to give rise to Anderson localization in disordered lattices. Incorporation of suitable gain media into these structures could enable self-optimized lasing from random nanocavities operating around the guided mode's cutoff, similar to what has been observed at the photonic band edge in *crescent-deviation* disordered PCs. It is worth noting that disordered waveguide structures could support self-optimized nanocavity lasers with significantly smaller modal volumes and lower thresholds than the large-area, disordered PC band-edge lasers.



(a) (b)

Figure 3 (a) Scanning electron microscope image of the fabricated Si-based two-dimensional membrane disordered photonic crystal microcavity; (b) calculated dispersion of the defect waveguide in ideal crystal shown in the inset (hollow circles denote odd modes and solid circles denote even modes).

The fabrication of the devices uses a simple scheme of incorporating colloidal PbSe QDs into the random PC microcavities. The disordered PCs were fabricated using the same procedure which is described in the previous chapter. A line-defect waveguide is formed by equally spaced circular holes defined in a hexagonal lattice of randomly rotated squares. The top image of the fabricated structure is shown in the SEM in Fig. 3(a). The thickness of the silicon slab (h=220 nm), the radius of the defect holes (r=105 nm), and the lattice constant (a=470 nm) and the fill factor (~30%) of the bulk PC were chosen so that the cutoff of the guided mode aligns spectrally with the PL peak of colloidal PbSe QDs at 1510 nm. The dispersion of the waveguide in the underlying periodic crystal calculated by plane-wave expansion method and the room temperature PL spectrum of the dots are shown in Fig. 3(b). The superimposed random scatterers which trigger mode-edge localization can be viewed as the difference between circles in the underlying (ideal) crystal and randomly oriented squares in the disordered crystal.

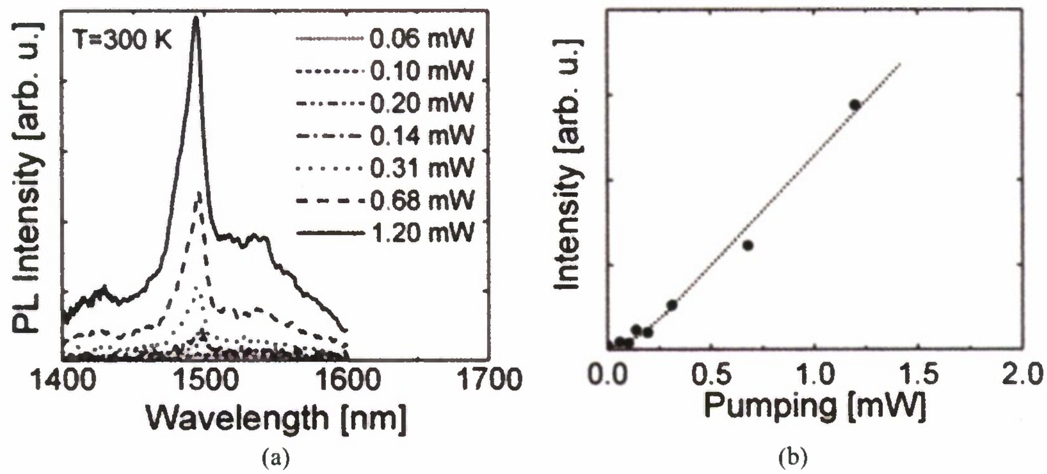


Figure 4 (a) Emission spectra of a silicon random photonic crystal microcavity with PbSe quantum dots measured at 300K at different pump powers; (b) light-light characteristics versus pump powers.

Colloidal PbSe QDs were embedded in the nanoscale air holes comprising the line defects in disordered PCs by soaking the samples in the PbSe QD solution. The devices were optically excited at room temperature with a cw Ti:sapphire laser operating at 810 nm. Emission from the QDs in the microcavities was focused by a high-resolution 100 × objective lens. The diffraction-limited size of the focus spot of the pump beam is ~1 μm. The localized modes have the minimum localization lengths of few lattice constants. The small focal spot allows us to efficiently pump these highly localized modes. It should also be mentioned that the disordered structure supports multiple spatially overlapping modes of various localization lengths at most probing frequencies. It means that, in the active structure, multiple modes are likely to be excited. Unlike emission from the conventional, engineered PC microcavities, the exact position of which is known, the output spectral characteristics for the random microcavities are sensitive to the location of the excitation. The disordered waveguides were probed systematically by scanning the focused pump beam along the waveguide axis. A strong dependence of excited modes' spectral characteristics on the excitation position was observed. Figure 4(a) shows a typical emission spectrum collected from a single excitation spot for varying excitation intensities. At lower pump intensity, the spectrum exhibits a broad spontaneous emission. Once the pump intensity exceeds a certain threshold, a much narrower emission peak emerges (~4 nm linewidth). It is possible that multiple random resonances are being excited according to the non-Lorentzian line shape of emission peak. There is a visible shoulder to the peak, and hence the linewidth of the emission peak is estimated by fitting the main peak without the shoulder. The plot of the peak emission intensity versus the pump intensity (L-L), shown in Fig. 4(b), exhibits a soft threshold at ~100 μW. The data shown in Fig. 4 do not indicate lasing, but suggest the onset of enhanced spontaneous emission coupled into localized modes as a result of strong feedback from random PC microcavities. Such feedback enables photon intensity around the resonance peak to quickly build up over that of the background luminescence.

3. Electroluminescence from silicon-based photonic crystal microcavities with Pbse quantum dots

We have demonstrated optically pumped silicon-based light sources using PbSe QDs embedded in various PC microcavities. However, for a more compact and versatile light source, an electrically injected laser is desired. To realize an electroluminescent device, it is necessary to link the QDs immersed in a conjugated polymer matrix with suitable charge transport layers. Additionally, the electroluminescence has to be coupled with a high-Q cavity for lasing.

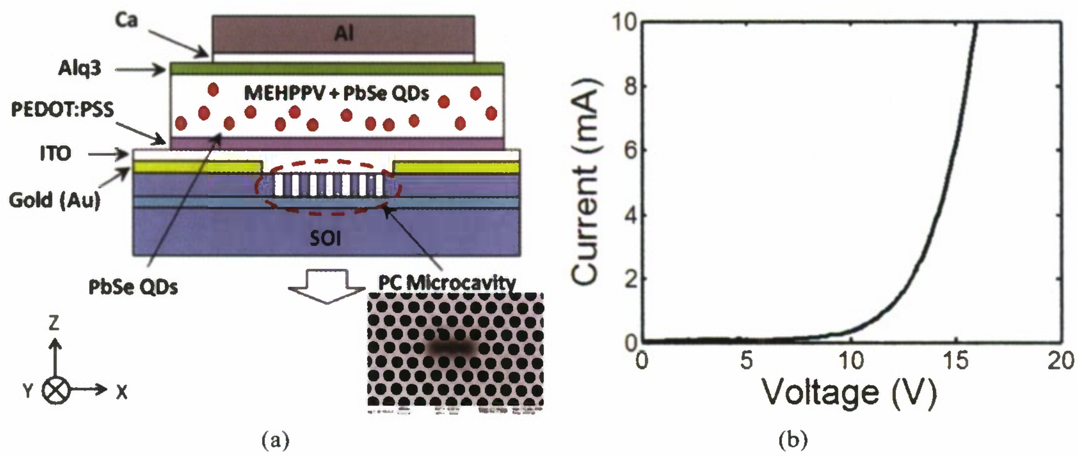


Figure 5 (a) Schematic of the device heterostructure fabricated on silicon-on-insulator (SOI). PbSe quantum dots with MEH-PPV are clad by PEDOT:PSS on the ITO anode and Alq3/Ca/Al cathode. The inset is a SEM image of the L3 defect photonic crystal microcavity in silicon. Outer air holes at both edges in the defect are shifted by 0.1*a*; (b) measured current-voltage characteristics of a fabricated device with a turn-on voltage of ~12 V.

In order to fabricate a device suitable for electrical injection, PbSe colloidal QDs are mixed with poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylene vinylene] (MEH-PPV), a conductive polymer matrix, and sandwiched between an indium tin oxide (ITO) anode and a thermally evaporated top cathode consisting of tris(8-hydroxyquinoline)aluminum (Alq3), calcium, and aluminum. The device heterostructure is schematically shown in Fig. 5(a). The ITO surface is altered by a layer of poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS), the purpose of which is two-fold: to make it smooth to prevent electrical shorts, and to align the Fermi level of ITO with the hole state of PbSe QDs as closely as possible for the efficient hole injection.

The resonator was designed as an L3 defect PC microcavity with three missing holes in a line in the silicon layer of a SOI wafer. The PC is therefore clad by SiO_2 and ITO on the two opposite surfaces. The outer air holes at both edges of the L3 cavity are shifted by $0.1a$ (The inset of Fig. 5(a)) in order to reduce leaky components of the in-plane electric field and attain a high Q-factor. Since ITO is deposited on the photonic crystal with air holes, it was expected that a small amount of filling of the holes would be unavoidable due to the conformal nature of the deposition. However, as observed by SEM, the degree of filling is extremely small. The Q-factor and modal volume of the L3 cavities, as estimated from three-dimensional FDTD calculations, are 2,000 and 0.14 μm^3

($\sim 1.6(\lambda_c/n)^3$), respectively. Asymmetric cladding by SiO_2 and ITO, both having refractive indices larger than air (n=1) results in a smaller Q-factor and larger modal volume than the values quoted above with air cladding.

The fabrication of the device was done by positioning the active gain medium on the PC microcavities. A 90 μm diameter circular active area is first defined by metal (20 Å NiCr, and 130 Å Au) lift-off to block the background light. A two-dimensional photonic crystal, consisting of a triangular lattice of air holes with period a=400 nm and hole radius r=110-130 nm, is fabricated within this active area in the 230 nm silicon layer of the SOI wafer by electron beam lithography and inductively coupled plasma reactive ion etching using a SiO_2 etch mask. Subsequent ITO deposition on the PC microcavity is followed by spin casting of PEDOT:PSS and baking at 150°C for 10 min. PbSe QDs/MEH-PPV with a 45wt% of QDs is spin casted on top of the PEDOT: PSS layer and annealed at 100°C for 30 min in an inert environment with both oxygen and water concentration lower than 5 ppm. After the device is cooled down to room temperature, 30 nm Alq3, 5 nm Ca, and 160 nm Al are thermally evaporated in succession.

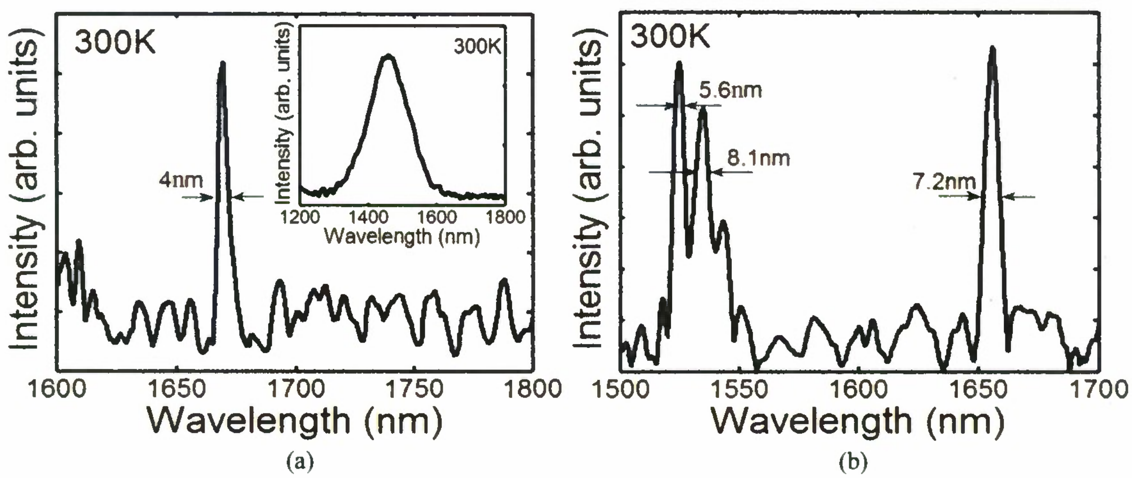


Figure 6 Room temperature electroluminescence spectra from different devices with silicon based PC microcavities with PbSe quantum dots. Resonant modes are observed at 1669 nm (a) with a linewidth of ~4 nm and at 1525 nm, 1535.6 nm, and 1656.2 nm (b) with the linewidths shown. The inset of (a) shows measured room temperature electroluminescence of the control device without a photonic crystal microcavity and a gold blocking layer. The device exhibits a broad emission wavelength between 1.3-1.65 μm with a linewidth of ~150 nm.

The devices were characterized at room temperature by applying a forward bias between the ITO anode and the Al cathode. Figure 5(b) shows the measured current-voltage (I-V) characteristics which exhibit diode-like behavior with a turn on voltage of ~12 V. The light output from the device was analyzed with a 0.75 m high-resolution spectrometer and detected with an InGaAs photomultiplier tube using phase lock-in amplification. A control device without the PC microcavity and gold blocking layer was also fabricated and characterized in order to measure the electroluminescence spectra of the PbSe QDs. The output spectra of this device exhibit a broad emission between 1.3-1.65 μm, with a full-width-at-half-maximum (FWHM) of ~150 nm (The inset of Fig. 6(a)). On the other hand, as shown in Figs. 6(a) and (b), the room temperature spectral output of devices with PC microcavities exhibit distinct resonances at wavelengths ranging from 1525 nm to 1669 nm at injection currents varying from 8-12 mA (a current density of 113-170 mA/cm^2). The narrowest linewidth observed is ~4 nm at λ=1669 nm, which corresponds to a cavity Q factor of ~420. We believe that the light from the active gain medium is not perfectly coupled and guided in the PC slab because of their spatial separation. This results in a lower value of Q than calculated. With an assumed dot density of 10^{12} /cm^2, ~10^6 carriers are injected into a PbSe QD per second, which implies that it would be impossible to attain stimulated emission. Therefore, the observed resonance is a manifestation of the Purcell effect which enhances the spontaneous emission. The maximum achievable Purcell factor is expressed as $F_C = 3\lambda_c^3 Q / 4\pi^2 n^3 V_{mode}$. With an estimated mode volume of ~$1.6(\lambda_c/n)^3$ and a cavity Q of ~420, we estimate F_C to be ~25 in the electroluminescent devices.

4. Plasmon Enhanced Nanolaser

Our objective is to demonstrate a coherent light source scalable to nanometer scale by means of photon confinement provided by the surface-bound plasmon wave. The effective index of the surface plasmon polariton (SPP) mode can have large effective index, thereby decreasing the resonance wavelength with respect to the cavity size. This can effectively reduce the cavity radiation loss. SPP mode requires TM gain, rather than typical TE, from InGaAs/GaAs quantum dots (QDs). This can be achieved by incorporating tensile-strained barrier in the growth of QDs.

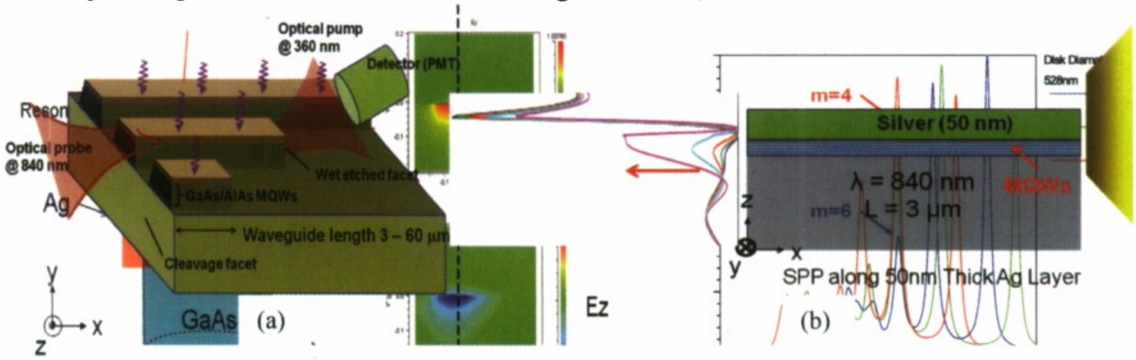


Figure 8 (a) Schematic of waveguide structure with Ag capping to generate SPP wave at the interface between Ag and GaAs MQWs; (b) FDTD simulation of enhanced transmission by MQW gain.

Figure 7 Simulation showing the SPP existing at the interface between Ag "hat" and GaAs substrate. Gains are provided by the quantum wells adjacent to the Ag film. Our simulation shows that the SPP wave are tightly confined at the interface. The penetration into the GaAs layer (~60 nm) sets a limit on the thickness of MQWs to be used in this structure.

The nanoscale laser utilizes the SPP wave established between the metal and semiconductor interface. FDTD simulation shows that the Whispering Gallery Modes (WGM) of the SPP wave have excellent scaling properties (Fig. 7(b)). Our calculations have shown that for the resonance in the NIR range, the E-field is TM dominant. Therefore it is preferred that the MQWs can provide TM gain. TM mode gain can be provided by tensile-strained quantum wells.

Critical to the success of the proposed nanolaser concept is to demonstrate that gain provided by the semiconductor quantum well can partially compensate the metal absorption loss of the SPP wave. Waveguide structures with Ag capping layer were fabricated to verify this. We first performed the FDTD simulation of the SPP propagation through such a waveguide structure (Fig. 8(a)). In this simulation, 50 nm thick Ag layer is used on top of GaAs substrate. It can be seen that the SPP fields are amplified at the Ag-GaAs interface with the aid of gain (Fig. 8(b)). The SPP amplitude at the air/Ag interface decrease with gain. We fabricated the waveguide structure to characterize the propagation of SPP wave with the assistance of gain provided by MQWs on GaAs substrate. Varying waveguide length allows direct measurement of SPP gain. This experiment was done in collaboration with colleague Prof. P.C. Ku. In this preliminary experiment, we observed that the transmission amplitude of the SPP wave was enhanced when the MQWs are optically pumped. And the increase in transmission follows that of the pumping power as shown in Fig. 9. This implies that round-trip gain is achievable in our proposed surface-plasmon enabled nanolaser structure.

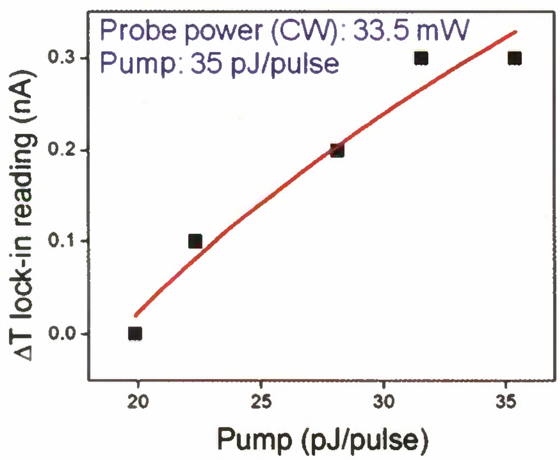


Figure 9 Measurement results showing enhanced transmission of SPP wave when the MQWs are optically pumped.

Conclusion:

Novel silicon based light emitters utilizing colloidal PbSe QDs which are incorporated in PC microcavities have been designed, fabricated, and characterized. We demonstrated two optically pumped devices which have different PC microcavities : the engineered and the self optimized random cavity. For an engineered H2 PC microcavity, efficient coupling of emission from PbSe QDs to silicon PC microcavity is achieved by inserting the QDs in a central air hole in the microcavity. Enhancement of spontaneous emission with a linewidth of ~2.0 nm, corresponding to a cavity Q factor of 775, is observed at 1550 nm at room temperature. We also demonstrated that random cavities in disordered PC waveguides with embedded colloidal PbSe QDs exhibit a peak emission at ~1.5 μm with a minimum linewidth of 4 nm. For more compact and versatile devices, we investigated and demonstrated enhanced spontaneous emission at room temperature from electrically injected silicon photonic crystal microcavities with PbSe QDs as the gain media. With a current density of 113 mA/cm^2, a resonance at λ=1669 nm having a linewidth of 4 nm is observed, which corresponds to a cavity Q factor of ~420 and a Purcell factor of ~25. This nanoscale light sources based on silicon, which is capable of being fabricated on CMOS chips, is of interest as a practical technology for optical interconnects in silicon photonics.

On the other hand, we have tried to demonstrate a coherent light source which can be scalable to sub-micron utilizing the SPP wave. We fabricated and characterized the waveguide structure along which the SPP wave can propagate with the assistance of gain provided by MQW. We demonstrated that round-trip gain in surface plasmon enabled nanolaser structure is achievable in the sense that the transmission in the waveguide increases as the pumping power increases.

Archival publications (published) during reporting period:

1. J. Heo, T. Zhu, J. Xu, and P. Bhattacharya, "Electroluminescence from silicon-based photonic crystal microcavities with PbSe quantum dots," *Appl. Phys. Lett.*, under review.

2. M. W. Kim, Y. H. Chen, J. Moore, Y. K. Wu, L. J. Guo, P. Bhattacharya, and P. C. Ku, "Sub-wavelength surface Plasmon optical cavity-Scaling, Amplification, and coherence," accepted to *IEEE J. Sel. Top. Quantum Electron.*, March 2009.

3. J. Yang, J. Heo, T. Zhu, J. Xu, J. Topolancik, F. Vollmer, R. Ilic, and P. Bhattacharya, "Enhanced photoluminescence from embedded PbSe colloidal quantum dots in silicon-based random photonic crystal microcavities," *Appl. Phys. Lett.* **92**, 261110 (2008).

4. Z. Wu, Z. Mi, and P. Bhattacharya, T. Zhu, and J. Xu, "Enhanced spontaneous emission at 1.55μm from colloidal PbSe quantum dots in a Si photonic crystal microcavity," *Appl. Phys. Lett.* **90**, 171105 (2007).

Conference Presentations:

1. J. Heo, T. Zhu, J. Xu, and P. Bhattacharya, "Electroluminescence from silicon-based photonic crystal microcavities with PbSe colloidal quantum dots," CLEO/IQEC 2009, Baltimore, MD, May 31-June 5, 2009.

2. M. W. Kim, J. Moore, Y. H. Chen, Y. K. Wu, P. Bhattacharya, L. J. Guo, and P. C. Ku, "Gain-assisted surface plasmon microcavity," CLEO/IQEC 2009, Baltimore, MD, May 31-June 5, 2009.

3. J. Yang, J. Heo, J. Xu, F. Vollmer, J. Topolancik, R. Ilic, and P. Bhattacharya, "Excitation of silicon-based random photonic crystal nanocavities with PbSe colloidal quantum dots," CLEO/QELS 2008, San Jose, CA, May 4-9, 2008.

4. M. W. Kim, J. Moore, J. Guo, P. Bhattacharya, and P. C. Ku, "Experimental characterization of gain assisted surface Plasmon propagation using quantum wells," IEEE/LEOS Annual Meeting 2008.

5. M. W. Kim, P. C. Ku, and J. Guo, "Surface Plasmon enabled subwavelength nano-cavity laser," IEEE/LEOS Annual Meeting 2007.

Awards and honors during reporting period:

1. Professor P. Bhattacharya was elected member of the National Academy of Engineering.
2. Professor P. Bhattacharya received the 2007 IEEE Nanotechnology Pioneer Award.
3. Professor P. Bhattacharya received the 2008 TMS John Bardeen Award.